

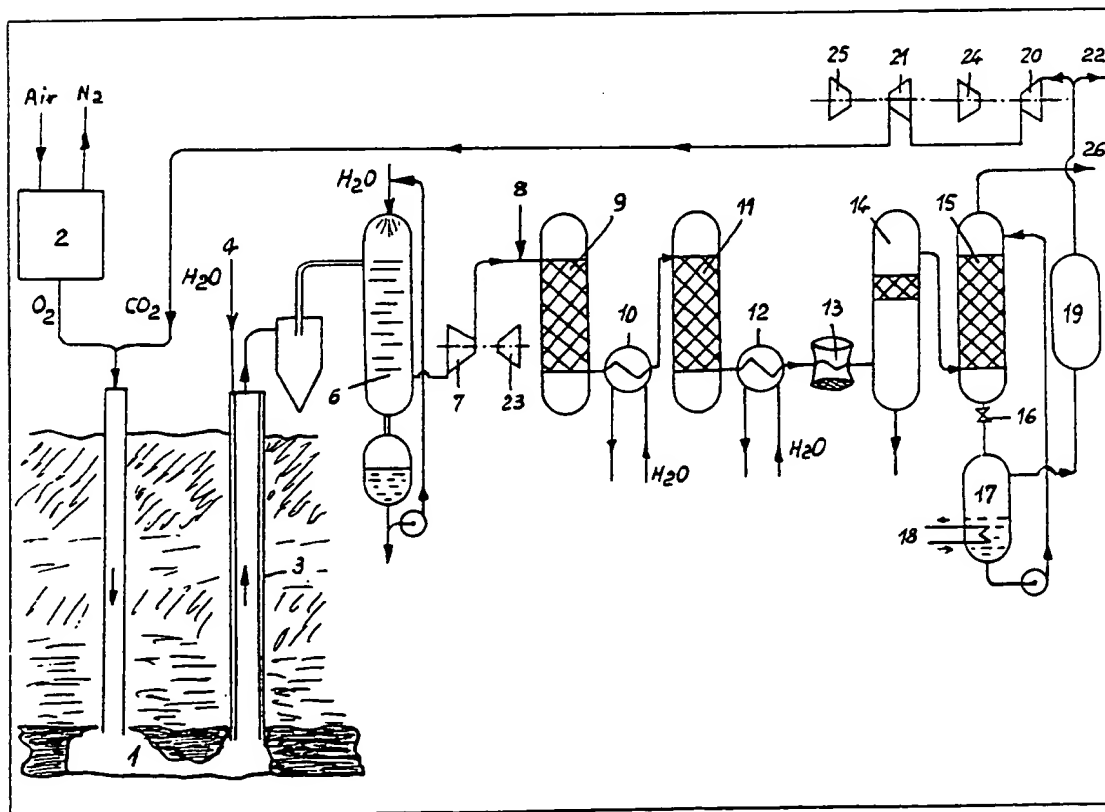
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(54) Method of producing a gas with a high hydrogen content by subterranean gasification of coal

(57) A method for the production of a hydrogen rich gas by underground gasification of coal, characterised in that it consists of associating an underground coal gasification operation carried out by means of

oxygen and CO_2 , the CO_2 being recovered in the plant in which the gas produced is cleansed, with an operation to cool the crude gas by the injection of water into the bores through which the gas producer discharges and an operation to convert the CO to CO_2 , carried out on the surface, employing the water vapour produced during cooling of the gas.

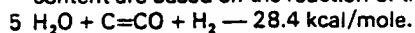


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SPECIFICATION

Method of producing a gas with a high hydrogen content by subterranean gasification of coal

All the extracted coal gasification processes which aim to produce a gas with a high hydrogen content are based on the reaction of the gas with water which can be expressed as:



Generally, the heat required for this reaction to take place is obtained by combustion of a portion of the batch.

In older processes, periodic inversions were carried out with the alternate injection of air and steam.

10 More recent processes employ continuous blowing using a gasifying agent consisting of a mixture of vapour and oxygen. 10

The same gasifying mixture has been used in the U.S.S.R. and in the U.S.A. during early attempts to produce a gas with a high hydrogen content performed by subterranean gasification of coal at a relatively low pressure in coal deposits located at depths no greater than 300 m.

15 If the production of a hydrogen rich gas by subterranean gasification of mineral coal deposits located at a greater depth is envisaged (beyond 700 or 800 m), the costing of the process makes it necessary to increase the gasification pressure to a minimum level of around 30 to 40 bars. 15

Under these conditions of working at great depth and under high pressure, the use of an oxygen vapour mixture as a gasifying agent in which the proportion of vapour may be as much as 65 to 85% is not without its drawbacks. 20

To avoid any risk of condensation of the vapour, the temperature of the mixture must at all points remain above a minimum level of around 250°C.

This high temperature makes it necessary to reduce the length of the lines used for injecting the gasifying agent and of inserting here and there expansion compensators. Under these conditions, it is virtually out of the question to use a gasification method in which the gasifying agent would be carried through galleries cut by conventional mining techniques and the injection of the gasifying agent by vertical bores providing direct access to the underground gas producer can only be carried out by means of relatively large diameter bores inside of which a heatproof tube is installed. 25

Underground gasification employing an oxygen vapour mixture entails two other types of drawback. 30

By reason of its relatively high temperature, the gasifying agent cannot preheat on contact with the rocks which surround the gas producer and it may, on the contrary, give off to them a part of its substantial heat which will proportionately reduce the efficiency of the gas producer in terms of energy.

Furthermore, in all underground gasification processes there is an interference between the performance of gasification reactions and the performance of coal pyrolysis reactions, the volatile substances which are released in the depths of the vein having no means of escape other than the gas-solid contact surface along which the gasification reactions take place. 35

The release of these volatile substances which contain considerable quantities of hydrogen has a marked inhibiting effect on the reaction of the gas with water; the result is a reduction in the degree of decomposition of the vapour injected and a degradation of the gasification efficiency. 40

The object of the process according to the invention is to remedy these various drawbacks.

To arrive at this end, the process combines an operation for underground gasification of coal which is carried out by means of a mixture of oxygen and CO₂, an operation to cool the crude gas by injection of water into the borings through which the gas producer discharges and a CO conversion operation carried out at the surface employing the steam produced during cooling of the gas. 45

The process according to the invention is likewise characterised by the fact that the CO₂ needed to carry out the underground gasification is recovered in the installation in which the gas produced is purified, and in that the reaction heat released during the course of the CO conversion operation is used for producing vapour which is employed in a condensation cycle in order to produce a part of the energy needed for operation of the plant. 50

The process according to the invention is described hereinafter with reference to a diagram in the appended drawings.

The underground gas producer 1 is supplied with a gasifying agent injected at ambient temperature and at elevated pressure (for example 40 bars to 50 bars). This gasifying agent consists of a mixture of oxygen originating from the plant 2 for separating oxygen from the air and CO₂ emanating from the factory which processes the gas produced. 55

The crude gas arrives at the discharge from the gas producer at a temperature of around 600 to 800°C.

It is discharged to the surface via one or more gas bores such as 3, each of which comprises a metal lining cemented into the soil and an inner metal tube suspended from the shaft head and freely expandable towards the bottom. 60

A line 4 provided with suitable regulating devices injects water under pressure into the annular space which separates the lining from the inner tube. This water evaporates on contact with the wall of the inner tube and the resultant vapour blends with the crude gas at the foot of the shaft.

The rate of flow of gasifying agent and the rate of flow of cooling water are so regulated that the mixture of crude gas and vapour produced by the cooling water arrives at the surface at a pressure of around 15 bars to 20 bars and at a temperature which is of the order of 200°C.

This crude gas and vapour mixture passes through a heatproof cyclone 5 which eliminates the major part of the entrained solids, then a washer 6 which saturates the gas with humidity and which eliminates the fine dust and condensable hydrocarbons. 5

After this first cleaning operation, the mixture of gas and vapour passes through a compressor 7 which raises its pressure to a level of around 40 bars to 50 bars and its temperature to the vicinity of 300°C. Additional water or vapour is added through the pipe 8 in order to adjust the temperature and the moisture content of the gas to the optimum level required by the operation for catalytic conversion of the CO. 10

The gas is enriched in hydrogen by conversion of the major part of the CO which it contains, according to the reaction: $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2 + 9.8 \text{ kcal/mole}$.

This reaction is carried out in reactors 9 and 11 which are provided with a catalyst capable of operating in the presence of sulphurous compounds (for example a cobalt molybdate based catalyst). Each of the two reactors is followed by one or more recuperating boilers such as 10 and 12 which employ the heat released by the conversion reaction in order to produce high pressure vapour (a pressure of around 40 to 50 bars). Alternatively, it is likewise possible to envisage regrouping conversion reactors and recuperation boilers by employing fluidised bed reactors cooled by water evaporating tubes disposed within the bed. 20

After final cooling of the gas in the cooler 13 and after elimination of the water and condensate in the separator 14, the gas is introduced into the reactor 15 in which separation of the major part of the CO_2 and H_2S is carried out by washing under pressure, employing a suitable solvent.

Still under pressure, the cleaned gas is discharged through the pipe 26 to be directed towards the chemical synthesis plant or to the distribution network. 25

The solvent which is expanded to atmospheric pressure in the pressure relieving apparatus 16 passes into the separator 17 in which the liquid is raised to the desired temperature by a heating circuit 18.

The CO_2 and the H_2S separate and are carried into the desulphuration plant 19 in which the major part of the H_2S is eliminated by conventional chemical technique. 30

The CO_2 needed for underground gasification is recompressed up to injection pressure in a multi-stage compression plant such as 20 and 21 with interspersed coolants. Excess CO_2 is eliminated via the pipe 22 so that it can be used for other purposes or be discharged into the atmosphere.

The various compressors are operated by steam turbines such as 23, 24 and 25 which are supplied with vapour produced in recuperation boilers 10 and 12. 35

The advantages which may be derived from replacement of a gasifying agent containing a considerable quantity of high temperature vapour by a gasifying agent consisting of a mixture of oxygen and carbon dioxide distributed at ambient temperature have already been pointed out hereinabove.

This replacement makes it possible to reduce the diameter and the cost of the gasifying agent injection bores; it also makes it possible to envisage the use of a combined method comprising a preparation of gasification sites by conventional mining techniques and a distribution of the gasifying agent through a network of pipes made in the underground galleries. 40

One can however wonder whether these advantages are not offset by a considerable reduction in efficiency from the energy point of view, the consequence of which would be a substantial increase in the prime cost of the gas produced. 45

To meet this objection, there follows a comparative examination of the production of gas with a high hydrogen content by the conventional gasification method employing an oxygen-vapour mixture and production of the same type of gas by the method which is the object of the present invention.

By way of example, the case chosen is one of a plant comprising an underground gas producer functioning at a pressure of 32 bars with a discharge temperature of 900°C and the object of which is to produce a gas intended for methanol synthesis, where the molar ratio of $\text{H}_2 : \text{CO}$ must be slightly above 2. The pressure at which the gasifying agent is injected is assumed to be 45 bars and the useful pressure of the gas produced 15 bars. 50

A calculation model is employed which is based on the conventional balances of $\text{H}_2\text{O} + \text{C}$ and $\text{CO}_2 + \text{C}$ reactions and on the hypothesis that the methane produced emanates substantially from decomposition of the volatile matter in the coal. 55

Applying this model to a deposit of anthracite-bearing coal containing 7% volatile matter in relation to pure coal, the following results are found:

I. Gasification by an oxygen-vapour mixture

Composition of the gas (% by volume of crude gas)

| | Gas | Crude at 900°C | Cooled to 200°C | Converted | Cleansed | |
|----|--------------------|-------------------|--------------------|-----------|----------|----|
| 5 | CO : | 33.5 | 33.5 | 23.0 | 23.0 | 5 |
| | CO ₂ : | 15.5 | 15.5 | 26.0 | — | |
| | H ₂ : | 36.0 | 36.0 | 46.5 | 46.5 | |
| | CH ₄ : | 2.5 | 2.5 | 2.5 | 2.5 | |
| | H ₂ O : | 12.5 | 62.5 | 52.0 | — | |
| 10 | | 100.0 | 150.0 | 150.0 | 72.0 | 10 |

Under these working conditions, the gasification efficiency (P.C.I. of the crude gas/P.C.I. of the gasified coal) is as much as 88%.

The consumption of gasifying agents amounts to:

- 0.175 mole of oxygen and
- 0.407 mole of vapour per mole of crude gas produced.

II. Gasification employing an oxygen-CO₂ mixture

Composition of the gas (in % by volume of crude gas)

| | Gas | Crude at 900°C | Cooled at 200°C | Converted | Cleansed | |
|----|--------------------|-------------------|--------------------|-----------|----------|----|
| 20 | CO : | 55.0 | 55.0 | 20.0 | 20.0 | 20 |
| | CO ₂ : | 37.0 | 37.0 | 72.0 | — | |
| | H ₂ : | 5.5 | 5.5 | 40.5 | 40.5 | |
| | CH ₄ : | 2.2 | 2.2 | 2.2 | 2.2 | |
| | H ₂ O : | 0.3 | 50.3 | 15.3 | — | |
| 25 | | 100.0 | 150.0 | 150.0 | 62.7 | 25 |

The gasification efficiency (P.C.I. of the crude gas/P.C.I. of the gasified coal) is 86%.

The consumption of gasifying agents amounts to:

- 0.187 mole of oxygen and
 - 0.441 mole of CO₂ per mole of crude gas produced.
- If these figures for gasification efficiency and consumption of gasifying agents are reduced to one and the same production of cleaned gas, the figures obtained are to the advantage of gasification employing the oxygen-vapour mixture.
- Indeed, taking into account the reduction in calorific output resulting from the CO conversion operation, we have:

35 For gasification using oxygen-vapour:

Gasification efficiency (cleaned gas/coal) = 86.1%

$$\text{Oxygen consumption: } 0.175 \times \frac{100}{72} = 0.243 \text{ mole/mole}$$

$$\text{Consumption of vapour: } 0.407 \times \frac{100}{72} = 0.565 \text{ mole/mole}$$

For gasification employing oxygen-carbon dioxide:

Gasification efficiency (cleaned gas/coal) = 79.3%

$$\text{Oxygen consumption: } 0.187 \times \frac{100}{62.7} = 0.298 \text{ mole/mole}$$

$$\text{CO}_2 \text{ consumption: } 0.441 \times \frac{100}{62.7} = 0.703 \text{ mole/mole}$$

- 5 However, the conclusion is amended if one examines the overall energy balance of the process; indeed, two major factors are to the advantage of the oxygen-CO₂ process: 5
- 1) the compression of one mole of CO₂ at 1 bar to 45 bars consumes substantially less energy than the production of one mole of vapour at the same pressure;
 - 2) the heat liberated by the operation to convert the CO is not heat lost, but heat which can be 10
- recovered in the form of vapour at a relatively high thermal level (of around 300 to 400°C). 10
- Bearing in mind these two factors and if one attributes an efficiency of 40% to the operation of conversion of heat energy into mechanical energy, the final energy balance looks like this:

Gasification using oxygen-vapour

| | kcal/cu.m. N clean gas | % of P.C.I. of gasified coal | |
|---|---------------------------|---------------------------------|----|
| 15 | | | 15 |
| Energy content of the gas: | 2,927 | 86.1 | |
| Heat recovery from conversion: | + 64 | + 1.9 | |
| Production of oxygen and vapour: | -630 | -18.5 | |
| Compression of the gas prior to conversion: | -200 | - 5.9 | |
| 20 | 2,161 | 63.6 | 20 |
| Energy efficiency | | | |

Gasification using oxygen-CO₂

| | kcal/cu.m. N clean gas | % of P.C.I. of gasified coal | |
|---|---------------------------|---------------------------------|----|
| | | | |
| Energy content of the gas: | 2,927 | 79.3 | |
| 25 | +246 | + 6.6 | 25 |
| Heat recovery from conversion: | | | |
| Production of oxygen and compression of CO ₂ : | -610 | -16.5 | |
| Compression of the gas prior to conversion: | -230 | - 6.2 | |
| Energy efficiency | 2,333 | 63.2 | |

- 30 These results show that from the point of view of energy efficiency the two processes are virtually the same and this conclusion would be further reinforced if separation of the CO₂ in the separator 17 were to take place at optimum pressure, in excess of atmospheric pressure, and if one were to take into account the inhibiting effect of the liberation of volatile matter from the coal on the gas reaction to water. 30

- 35 To sum up, the process according to the invention can benefit from all the advantages made available by employing a non-condensable gasifying agent which can be used at ambient temperature without involving any disadvantages with regard to the energy efficiency of the plant. This result is obtained by an association of the operations of underground gasification of the coal, cooling of the gas and conversion of the CO which makes it possible chemically to exploit the inevitable vapour produced by cooling of the crude gas under pressure and temperature conditions which are too low to allow it to be used to advantage in a thermodynamic cycle and which makes it possible to exploit the heat released 40 by the conversion reaction, producing steam at a relatively high temperature which can be employed in a condensation cycle to produce some of the energy needed for the plant to operate. 40

The energy economics of the process are likewise favoured by the fact that the CO₂ which results from cleansing of the gas can be employed as a gasifying agent and that the compression of the CO₂ consumes less energy than the production of an equivalent quantity of water vapour or steam.

CLAIMS

- 5 1. A method for the production of a hydrogen rich gas by underground gasification of coal, 5
characterised in that it consists of associating an underground coal gasification operation carried out by
means of oxygen and CO₂, the CO₂ being recovered in the plant in which the gas produced is cleansed,
an operation to cool the crude gas by the injection of water into the bores through which the gas
producer discharges and an operation to convert the CO₂, carried out on the surface, employing the
10 water vapour produced during cooling of the gas. 10
2. A method for producing a hydrogen rich gas according to Claim 1, characterised in that the
reaction heat released during the course of the CO conversion operation is used for producing the
vapour which is employed in a condensation cycle in order to produce some of the energy required for
operation of the plant.

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